

**EVIDENCE FOR A SINGLE STELLAR ENVIRONMENT OF R-PROCESS NUCLEOSYNTHESIS FROM LIVE  $^{247}\text{Cm}$  IN THE EARLY SOLAR SYSTEM.** F. L. H Tissot<sup>1,2</sup>, N. Dauphas<sup>1</sup>, L. Grossman<sup>3</sup>, <sup>1</sup>Origins Lab, Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, IL ([ftissot@uchicago.edu](mailto:ftissot@uchicago.edu)), <sup>2</sup>Department of the Earth, Atmospheric and Planetary Sciences, MIT, Cambridge, MA, <sup>3</sup>Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, IL.

**Motivation:** The Early Solar System (ESS) abundance of short-lived radionuclides (SLR) can be interpreted in terms of a free-decay interval ( $\Delta$ ) or a mixing timescale ( $\tau$ ), which correspond to complete or partial isolation from fresh nucleosynthetic inputs of the molecular cloud core parental to the solar system. For  $r$ -nuclides, however, the abundances measured in ESS materials require less isolation for  $^{107}\text{Pd}$  and  $^{182}\text{Hf}$  ( $\Delta \sim 35$  Myr) than for  $^{129}\text{I}$  ( $\Delta=100\pm 7$  Myr) and  $^{244}\text{Pu}$  ( $\Delta=158\pm 85$  Myr) [1]. To make sense of these observations, models have proposed the existence of up to three different  $r$ -processes producing, respectively, the light  $r$ -nuclides, the heavy  $r$ -nuclides and the actinides [1-4]. In this work [5], we determined the ESS abundance of another short-lived  $r$ -nuclide, the long sought after  $^{247}\text{Cm}$  [e.g., 6-7], and show how this additional constraint is consistent with either three different  $r$ -processes, or, and more likely, a single stellar environment of  $r$ -process nucleosynthesis and a partial  $s$ -process origin for  $^{107}\text{Pd}$  and  $^{182}\text{Hf}$ .

**Background:** Elements with mass above  $\sim 70$  amu are the products of three main processes of stellar nucleosynthesis: the  $s$ - (*slow* neutron capture),  $r$ - (*rapid* neutron capture) and  $p$ - (*proton*) process [8-9]. If the  $s$ - and  $p$ -processes are relatively well understood (neutron capture in AGB-stars and photodisintegration of seed nuclei in supernovae, respectively [10-11]), little is known regarding the astrophysical conditions under which  $r$ -process nuclides are produced [1-4].

In particular, the number of different  $r$ -processes is a topic of intense debate. Evidence for the existence of multiple  $r$ -processes comes from both spectroscopic observations of Low Metallicity Halo Stars (LMHS) and the relative abundance of SLR in meteorites. LMHS are stars with low metal content that formed early in the history of the Galaxy, before the onset of the  $s$ -process in AGB stars. Evidence for multiple  $r$ -processes in LMHS include: (i) the favored production of lighter  $r$ -nuclides (such as Sr) over heavier  $r$ -nuclides (such as Ba) ([12] and refs. therein); (ii) the obvious decoupling between  $r$ -nuclides of lower and higher atomic masses in the composition of star CS 22892-052 [13-14]; (iii) the overabundance of Th and U relative to lighter  $r$ -nuclides (e.g., Eu) in CS 31082-001 [15-16]; or even (iv) the gradual decrease in the abundance pattern of  $r$ -nuclides as a function of atomic number, from Sr to Yb, in star HD 122563 [17-20].

The second line of evidence pointing to a multiplicity of  $r$ -processes comes from the relative abundance of SLR in meteorites. In particular, the abundance of  $^{182}\text{Hf}$  ( $t_{1/2}=8.90$  Myr) in the ESS is too high relative to that of another short-lived  $r$ -nuclide,  $^{129}\text{I}$  ( $t_{1/2}=15.7$  Myr). This led Wasserburg et al. (1996) [2] to propose that different  $r$ -processes were producing the light  $r$ -nuclides ( $A\leq 135-140$ , up to  $\sim \text{Ba}$ ) and the heavy  $r$ -nuclides ( $A\geq 135-140$ ), respectively. The existence of an actinide-specific (e.g., U, Th, Pu) site was also hypothesized based on the low meteoritic abundance of  $^{244}\text{Pu}$  ( $t_{1/2}=79.3$  Myr) compared to  $^{182}\text{Hf}$  [1].

The existence of an actinide-specific site is, however, not necessarily warranted because  $^{244}\text{Pu}$  has a long half-life and its stellar yield is uncertain [21], which makes it poorly sensitive to the history of nucleosynthesis prior to SS formation and whether or not multiple  $r$ -process sites contributed to the synthesis of this nuclide. In contrast, Curium-247, which ultimately decays into  $^{235}\text{U}$ , has a much shorter half-life ( $t_{1/2}=15.6$  Myr) and is very well suited to address this question, provided its abundance in the ESS can be accurately and precisely determined.

**$^{247}\text{Cm}$  ESS abundance:** Evidence for  $^{247}\text{Cm}$  decay can only be found as  $^{235}\text{U}$  excesses in ESS materials. Yet, the ESS abundance of  $^{247}\text{Cm}$  expected from models of Galactic Chemical Evolution (GCE) is low, and a  $^{235}\text{U}$  excess from curium decay will therefore only be resolvable in phases which formed early in the SS and preferentially incorporated Cm over U. Recently,  $^{235}\text{U}$  excesses of up to +3.5 % (rel. to CRM-112a) were found in four fine-grained CAIs [7]. Those excesses correlate with Nd/U ratio (Nd is used a proxy for Cm, which has no stable isotope) and the authors interpreted their results as evidence for live  $^{247}\text{Cm}$  in the ESS.

A complicating factor is that  $^{235}\text{U}$  excesses of such magnitude could have also been produced by isotopic fractionation during evaporation/condensation processes in the early solar nebula. Indeed, the kinetic theory of gases predicts that the lighter isotope will condense (evaporate) faster during condensation (evaporation), leading to isotopic fractionation that scale as the square root of the mass of the reacting species: in the case of monatomic U in a low pressure gas,  $\alpha \sim \sqrt{238/235}-1 \sim 6.3$  %. The excesses found by [7] are smaller than 6 % and the question thus remained as to what were the causes of the observed U isotope variations.

In the present work [5], we conducted a survey of the U isotope composition in 12 fine-grained CAIs characterized by group II Rare Earth Element (REE) patterns, in which the light REE are enriched relative to the heavy REE and U. As Cm is expected to behave like a light REE during evaporation/condensation processes, group II CAIs, which display high Nd/U ratios (a proxy for the Cm/U ratio), are the best phases to investigate when looking for  $^{235}\text{U}$  excesses due to  $^{247}\text{Cm}$  decay. As in previous studies, most samples display  $^{144}\text{Nd}/^{238}\text{U}$  ratios below 600 and  $\delta^{235}\text{U}$  values within 6 ‰ of the bulk SS value. The samples display a trend between  $\delta^{235}\text{U}$  and  $^{144}\text{Nd}/^{238}\text{U}$  similar to that observed by [7], but there is significant scatter around the best-fit line. One sample (named *Curious Marie*) has an extremely high  $^{144}\text{Nd}/^{238}\text{U}$  ratio ( $\sim 13,720$ ) and a  $^{235}\text{U}$  excess of  $\sim +59$  ‰. Considerable effort was expended to confirm this result that was eventually triplicated using different analytical protocols, demonstrating that the  $^{235}\text{U}$  excess in *Curious Marie* is real.

Using the slope of the isochron ( $\delta^{235}\text{U}$  vs.  $^{144}\text{Nd}/^{238}\text{U}$  diagram), the  $(^{247}\text{Cm}/^{235}\text{U})_{\text{Initial}}$  ratio can be calculated. Because the slope of the isochron is mainly leveraged by *Curious Marie*, the initial  $^{247}\text{Cm}/^{235}\text{U}$  ratio obtained will correspond to the time when *Curious Marie* acquired its Nd/U ratio. The extreme U depletion observed in *Curious Marie* is likely due to solar nebula condensation and/or nebular/parent body alteration. The two can be disentangled by looking at the relative depletion of two refractory lithophile elements with similar volatilities (*i.e.*, expected to condense together during nebular processes). In most samples, U and Yb present similar levels of depletion relative to solar composition and the abundance of other refractory lithophile elements (*e.g.*, Nd or Sm), indicating that U and Yb have similar behaviors during evaporation/condensation processes under solar nebula conditions. In *Curious Marie*, however, the U/Nd ratio is  $1000\times$  lower than solar, while the Yb/Nd ratio is only depleted by a factor of 50. The twentyfold greater depletion in U relative to Yb in *Curious Marie* is likely due to alteration, either in the early nebula or on the parent body of Allende. Dating of aqueous alteration products on meteorite parent-bodies with extinct radionuclides  $^{36}\text{Cl}$ ,  $^{26}\text{Al}$ ,  $^{53}\text{Mn}$  or  $^{129}\text{I}$  suggests it took place no later than 10 Myr after SS formation [22-23]. Accounting for an alteration age of  $5\pm 5$  Myr, the  $(^{247}\text{Cm}/^{235}\text{U})_{\text{Initial}}$  ratio obtained from the two-point isochron between *Curious Marie* and average SS materials can be corrected to obtain a  $(^{247}\text{Cm}/^{235}\text{U})_{\text{ESS}}$  ratio of  $(1.1 \pm 0.3) \times 10^{-4}$ . This value is in agreement with the value of  $(1.1-2.4) \times 10^{-4}$  obtained by [7] based on CAI measurements and an upper limit of  $\sim 4 \times 10^{-3}$  inferred

from earlier meteoritic measurements [6]. It is also in line with the lower limit derived from GCE modeling:  $(5.0 \pm 2.5) \times 10^{-5}$  [21].

**Implication for the *r*-process:** Using the  $(^{247}\text{Cm}/^{235}\text{U})_{\text{ESS}}$  ratio determined above and the open non-linear GCE model of [24], a free-decay interval of  $\Delta = 87 \pm 14$  Myr is obtained. This value is in good agreement with the  $\Delta$  value of  $\sim 100$  Myr derived from  $^{129}\text{I}$  and  $^{244}\text{Pu}$  but is much longer than the value of  $\sim 35$  Myr obtained from  $^{107}\text{Pd}$  and  $^{182}\text{Hf}$ . Recent nucleosynthetic models have, however, reconsidered the origin of  $^{107}\text{Pd}$  and  $^{182}\text{Hf}$  and find a significant *s*-process contribution ( $\sim 80\%$ ) for both isotopes [3, 25]. In such a framework, the initial abundance of all *r*-process SLR in the ESS can be explained by a single *r*-process event, which last injected material into the protosolar molecular cloud  $\sim 100$  Myr before SS formation.

Our results indicate that the multiplicity of *r*-processes inferred from LMHS abundance patterns may only be relevant to exotic conditions that prevailed in the earliest generation of stars of the Galaxy and that a single *r*-process may still be relevant to long-term GCE models.

**References:** [1] Dauphas N. (2005) *Nucl. Phys. A* 758, 757C-760C. [2] Wasserburg G. J. et al. (1996) *APJ* 466, L109-L113. [3] Meyer B. S. and Clayton D. D. (2000) *Space Sci. Rev.* 92, 133-152. [4] Huss G. R. et al. (2009) *GCA* 73, 4922-4945. [5] Tissot F. L. H. et al. (Accepted) *Science Advances*. [6] Chen J. H. and Wasserburg G. J. (1981) *EPSL* 52, 1-15. [7] Brennecka G. et al. (2010) *Science* 327, 449-451. [8] Burbidge E. M. et al. (1957) *Rev. Modern Phys.* 29, 547-650. [9] Cameron A.G.W. (1957), *Stellar evol. nucl. astrophys. nucleogenesis*. [10] Bisterzo S. et al. (2014) *APJ* 787. [11] Rauscher T. et al. (2013) *Rep. Prog. Phys.* 76. [12] Truran J. W. et al. (2002) *Astron. Soc. Pacific* 114(802): 1293-1308. [13] Sneden C. et al (2003) *APJ* 591(2): 936. [14] Cowan J. J and Sneden, C. (2004) *Carnegie Observ. Astrophys. Series* 4, 1:27. [15] Cayrel R. et al. (2001) *Nature* 409, 691-692. [16] Hill, V. et al. (2002) *A&A*. 387, 560-579. [17] Sneden C. and Parthasarathy M. (1983) *APJ* 267, 757-778. [18] Westin J. et al (2000) *APJ* 530, 783. [19] Sneden C. and Cowan J. J. (2003) *Science* 299, 70-75. [20] Honda S. et al. (2006) *APJ* 643, 1180. [21] Nittler L. R. and Dauphas N. (2006) *Meteo. Chem. Evol. Milky Way* 127-146. [22] Ross D. K. et al (2015) *LPSC XLVI*, #2552. [23] Russell S. S. and MacPherson G. J. (1997) *Workshop parent-body nebular modif. chondr. materials*. 4054. [24] Dauphas N. et al. (2003) *Nuc. Phys. A* 719, 287C-295C. [25] Lugaro M. et al. (2014) *Science* 345, 650-653.